

A Kondo lattice antiferromagnet CePd_5Al_2

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We report on the electrical resistivity, magnetic susceptibility and heat-capacity measurements on a new intermetallic compound CePd_5Al_2 , crystallizing in the ZrNi_2Al_5 -type tetragonal structure, with lattice parameters $a = 4.156 \text{ \AA}$ and $c = 14.883 \text{ \AA}$. The compound presents Kondo lattice behavior and an easy-plane antiferromagnetic ground state with two magnetic transitions at 2.9 K and 3.9 K. The Sommerfeld coefficient is estimated as 60 mJ/mol K^2 .

KEYWORDS: cerium palladium aluminum, heavy fermion, Kondo lattice, antiferromagnetism

Cerium-based intermetallic compounds have been intensively studied due to the exceptional variety of physical properties they can display, and their potential to be used as model systems for understanding fundamental phenomena such as quantum critical points or unconventional superconductivity.¹ For example, among several compounds in the ternary system Ce-Ni-Al, CeNi_2Al_5 and CeNiAl_4 have attracted attention due to their interesting electric and magnetic properties.^{2–8} In fact, CeNi_2Al_5 is a heavy fermion compound with a magnetic transition at 2.6 K and a Kondo temperature $T_K \simeq 5 \text{ K}$. On the other hand, CeNiAl_4 is a non-magnetic heavy fermion compound with $T_K \simeq 67 \text{ K}$. In the Ce-Pd-Al system, the hexagonal compounds CePdAl and CePd_2Al_3 order antiferromagnetically below 2.7 K and 2.8 K, respectively. In the latter, a large Sommerfeld coefficient $\gamma = 340 \text{ mJ/mol K}^2$ was observed.^{9–11} Recently, a new transuranic compound NpPd_5Al_2 has been reported,¹² a rare example of a Np compound without magnetic ordering, which also presents a superconducting transition temperature at $T_C = 4.9 \text{ K}$. Motivated by these works, it seemed worthwhile to investigate the existence of a new Ce compound CePd_5Al_2 , which we have succeeded to prepare and herein report the physical properties.

A polycrystalline sample of CePd_5Al_2 was prepared by arc melting in a high purity argon atmosphere. The initial mixture was made directly from the elements (Ce: 99.9%, Pd: 99.99%, Al: 99.999%) in stoichiometric proportion and melted into a button. After that, it was sealed in an evacuated quartz tube and annealed at 800°C for 7 days. The chemical composition of the

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annealed button was evaluated by electron-probe microanalysis (EPMA) using a wavelength dispersive JEOL JXA-8200 system. A stoichiometric 1:5:2 proportion in the CePd_5Al_2 sample was confirmed and no secondary phases were found. Differential thermal analysis up to 1550°C revealed a single exothermal event at $1080 \pm 10^\circ\text{C}$, associated the compound's melting point. Powder x-ray diffraction also revealed that the sample presents a single phase, as shown in Fig. 1. The phase is isostructural to NpPd_5Al_2 : a tetragonal ZrNi_2Al_5 -type structure, with space group $I4/mmm$. The lattice constants obtained by refining the x-ray pattern in the Powder Cell program are $a = 4.156 \text{ \AA}$ and $c = 14.883 \text{ \AA}$. We could also observe in metallographic examination that the grains had an elongated shape of 1 mm in length and $100 \mu\text{m}$ in diameter, indicative of strongly preferential alignment. An x-ray diffraction pattern (Fig. 1.b) from the polished bottom of the sample showed high intensities for the $00l$ peaks, indicating that the c -axis is preferentially aligned with the long direction of the grains. A sample of LaPd_5Al_2 was prepared by the same method as for Ce compound, in attempt to obtain a non-magnetic reference compound. However, multiple phases were observable in the powder x-ray diffraction so this reference sample was not used in this work.

Based on the preferential alignment, elongated bar shaped samples were cut from the CePd_5Al_2 pellet parallel and perpendicular to the grain length for resistivity experiments. In addition, a cube shaped sample was cut for magnetization experiments parallel and perpendicular to this direction, and a platelike sample was cut for heat capacity experiments. The electrical resistivity $\rho(T)$ was measured by a AC four-probe method in two different systems: a ^3He refrigerator for temperatures between 0.3 K to 100 K, and a GM refrigerator home-built setup for 3 K to 400 K. The magnetization $M(T, H)$ was measured by a SQUID magnetometer (Quantum Design MPMS) from 2 to 350 K. The specific heat $C(T)$ was also measured using a Quantum Design PPMS, between 0.3 K and 300 K.

Fig. 2 shows the temperature dependence of electrical resistivity, $\rho(T)$, of CePd_5Al_2 with the current parallel and perpendicular to the grain alignment. The sample shows anisotropy and classic strongly correlated electron metal behavior¹³ with $\rho_{\parallel}(300 \text{ K}) = 24 \mu\Omega \text{ cm}$ and $\rho_{\perp}(300 \text{ K}) = 42 \mu\Omega \text{ cm}$, decreasing as a broad shoulder upon cooling until a broad local minimum is reached at 13 K. Kondo lattice behavior is then clearly seen (inset of Fig. 2), where a $-\ln T$ dependence appears between 10 K and 4 K. A drop in the $\rho(T)$ at $T_1 = 3.9 \text{ K}$ is ascribable to the reduction of the scattering of conduction electrons due the onset of Ce magnetic moments coherence and/or alignment. A second kink in the slope at $T_2 = 2.9 \text{ K}$ is also seen, marking a probable change in magnetic structure. Fermi-liquid behavior can be observed below 0.8 K and a $\rho = \rho_0 + AT^2$ fit below this temperature gives residual resistivity $\rho_0 = 1.37 \mu\Omega \text{ cm}$ and $A = 0.04 \mu\Omega \text{ cm/K}^2$. The residual resistivity ratio ($RRR = \rho(300 \text{ K})/\rho(0 \text{ K})$) is 18, which demonstrates a good quality of the sample.

The temperature dependence of the reciprocal susceptibility, $1/\chi = B/M$ at applied field

$B = 0.1$ T is shown in Fig. 3. Since the CePd_5Al_2 polycrystalline sample presented grain alignment, we could perform magnetization measurements in both directions, parallel and perpendicular to the grain length, corresponding to the solid and open circles in Fig. 3, respectively. The measurements indicate the presence of easy-plane magnetocrystalline anisotropy, with larger response perpendicular to the alignment of the grains, therefore to the c -axis. Curie-Weiss law fittings between 175 K and 275 K of the two datasets are shown as straight lines in the figure. For χ_{\parallel} , the Curie temperature is $\theta_{\parallel} = -59$ K, while for χ_{\perp} we obtained $\theta_{\perp} = -8.1$ K. The estimated effective moments are very similar, $\mu_{eff} = 2.53 \mu_B/\text{f.u.}$ and $2.56 \mu_B/\text{f.u.}$ respectively, indicating that in CePd_5Al_2 the Ce ions are trivalent. The large difference in θ suggests a crystalline electric field (CEF) splitting of the Ce ground state, and from the CEF model¹⁴ we can roughly estimate the value of $B_2^0 = \{[10(\theta_{\perp} - \theta_{\parallel})]/[3(2J-1)(2J+3)]\}$ as 5.3 K, but a single crystal will be necessary to do a more precise evaluation. In both directions, an antiferromagnetic transition with $T_{N1} = 3.9$ K is found, followed by a second magnetic transition at $T_{N2} = 2.9$ K. The inset in Fig. 3 shows the magnetization isotherms at 1.8 K for the parallel and perpendicular directions of the grains, revealing a metamagnetic behavior at $B \simeq 0.9$ T.

The specific heat $C(T)$ measured at zero field is presented in Fig. 4. Two very distinct peaks are seen, giving $T_{N1} = 3.9$ K and $T_{N2} = 2.9$ K, consistent with the double magnetic transitions found in resistivity and magnetization data. By extrapolating a linear behavior in C/T versus T^2 data between 8 K and 20 K (not shown) the Sommerfeld coefficient of CePd_5Al_2 is estimated as $\gamma = 60$ mJ/mol K². This not only provides information about the density of states at the Fermi level for a metal, it is also a good parameter to evaluate the strength of electronic correlations. In a classical metal γ is typically rather small (~ 10 mJ/mol K²), while true heavy-fermion systems have values higher than 400 mJ/mol K. In CePd_5Al_2 the intermediate γ value indicates that electronic correlations are present, but it cannot be considered a true heavy-fermion compound. Integration of a C/T versus T plot gives an estimation of the total entropy S (inset of Fig. 4) which includes magnetic, electronic and lattice contributions. The total entropy of CePd_5Al_2 does not reach $R \ln 2$ at the T_{N1} , and does only well above the magnetic transitions, at $T \approx 13$ K. This is further indication of electron correlations and magnetic screening effects, i.e., the $4f$ electrons of the Ce ions do not behave as simple localized electrons. The CEF ground state of the Ce ions should be a doublet, similar to the CeNi_2Al_5 compound.¹² The Kadowaki-Woods ratio A/γ^2 results in the expected value of $10^{-5} \Omega \text{ cm (mol K/J)}^2$, also supporting a ground-state doublet.^{15,16}

In summary, we have prepared a new compound CePd_5Al_2 , which presents two magnetic transitions at $T_{N1} = 3.9$ K and $T_{N2} = 2.9$ K. The resistivity measurements reveal Kondo lattice and Fermi-liquid behavior. The compound has anisotropic magnetization and presents a CEF splitting of the Ce ground state, therefore we are now making efforts to grow single

crystals for further investigations.

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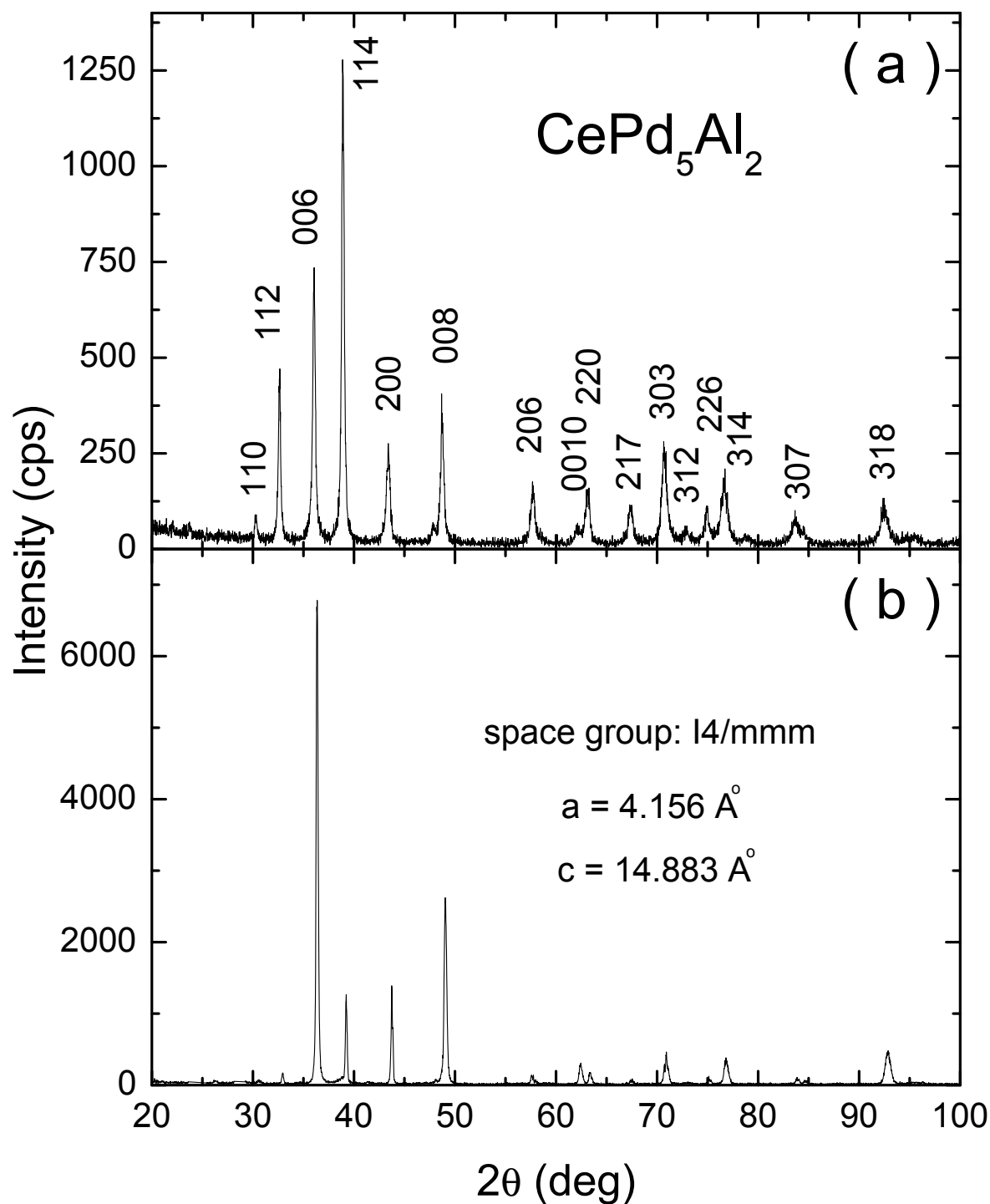


Fig. 1. X-ray diffraction ($\text{Cu } K\alpha$) pattern of CePd_5Al_2 at room temperature. (a) powdered sample, (b) bottom surface of the arc melted button.

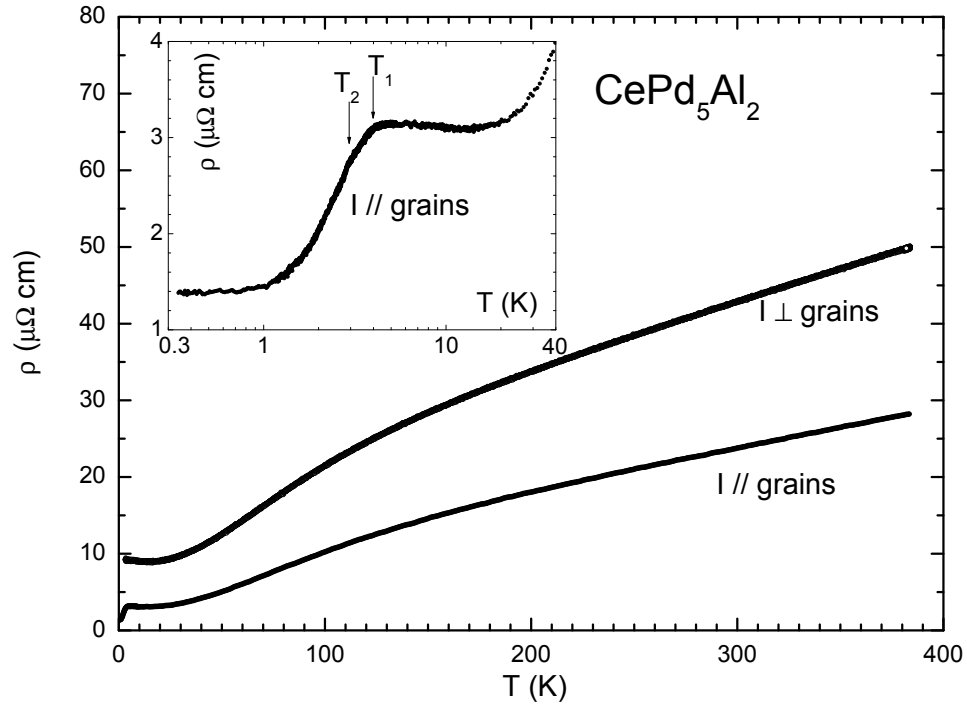


Fig. 2. Temperature dependence of the electrical resistivity, $\rho(T)$, of CePd_5Al_2 . The inset shows the $\rho(T)$ vs $\ln T$ plot and the arrows mark the two anomalies.

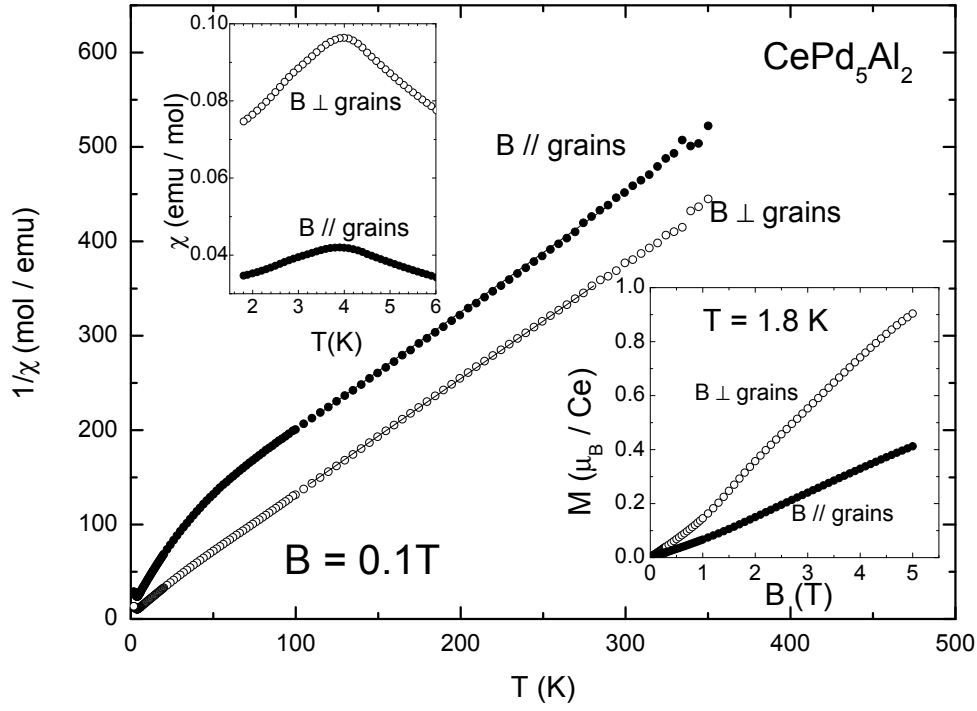


Fig. 3. Temperature dependence of the inverse magnetic susceptibility $1/\chi = B/M$ for CePd_5Al_2 at $B = 0.1$ T, showing magnetocrystalline anisotropy and Curie-Weiss behavior at high temperatures. The solid circles and open circles are the measurements with B parallel and perpendicular to the grain lengths, respectively. The upper inset shows the low temperatures data of $\chi(T)$. The lower inset shows the magnetization isotherms at $T = 1.8$ K, revealing a metamagnetic behavior at 0.9 T for both orientations.

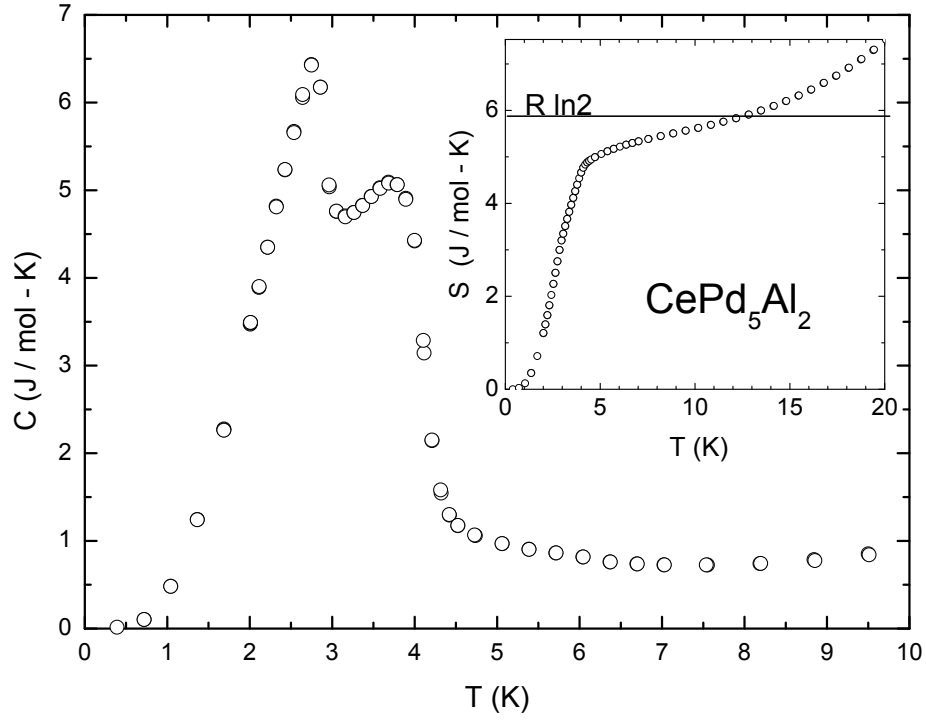


Fig. 4. Temperature dependence of the specific heat $C(T)$ for CePd_5Al_2 , showing a double peak structure at low temperatures. The inset shows the total entropy $S(T)$.